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Atmospheric Measurement Techniques Discussions

Interactive comment on "Characterizing the Arctic absorbing aerosol with multi-instrument observations" *by* Eija Asmi et al.

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We wish to thank the reviewer for their excellent comments and suggestions that have greatly helped us to re-structure and improve the presentation of our campaign results in the form of this manuscript. We hope that the reviewers find the changes acceptable and the current version of the manuscript more appropriate for publication in AMT. We have modified the manuscript content significantly. We summarize below the major changes that were done and answer the detailed reviewers' comments below the summary.

Major changes, summary:

The modified manuscript focuses entirely on instruments capabilities for detecting low concentrations in a pristine field environment. All discussion on atmospheric relevance and implications of the measured aerosol concentrations, including the analysis of air mass origin and Arctic BC mixing state were removed. All figures, tables and instrument correlations were re-calculated using 100% of campaign data (i.e. not separately for periods 1, 2 or neglecting some periods). This affected slightly the correlation statistics described in subsection 3.4 (current 3.3). Results section structure was modified such that the former subsection 3.3 (on detection limits) was moved at the beginning of section 3 (current 3.1) and Allan variances were calculated and used to determine the lowest averaging times of the instruments. Subsection 3.1 is now followed by modified and compressed former sections 3.1 and 3.2 (campaign overview and observed absorption values). The former section 3.5 (Particle size and coating impact on measured absorption) was removed completely and a new short section 3.4 with a simplistic MAC-value calculation was added. The main conclusions of the modified manuscript are: - filter-based methods are sensitive to detect absorption coefficients down to around 0.01 Mm-1 level (1-sigma) while the use of EMS method requires around 10-fold higher absorption coefficient values - Arctic summer absorption values were most of the time between 0.06-0.1 Mm-1 in our study, which is well above the lowest detection limits of filter-based instruments, but too low for EMS methods - Even at these low concentrations, the absorption values measured by different filter-based instruments show a good linear correlation, confirming both the accuracy and the precision seem to be adequate. Here the exception was the absorption measured by COSMOS instrument, where the pre-treatment of the sample can, as expected, modify the measured absorption. This led to a slope clearly below 1 between MAAP and COSMOS absorption. - Mass-absorption cross section of 16 m2 g-1 was determined for MAAP, when compared to a residual BC mass measured by SP2. This is well in-line with the MAC values obtained in a comprehensive study in the Arctic by Ohata et al., 2020.

Following references were added: Werle et al., 1993; Ohata et al., 2020; Allan et al.,

1966; Hagler et al., 2011; Springston and Sedlacek, 2007; Laing et al., 2020; Modini et al., 2021; Torseth et al., 2019; Bond et al., 2006; Jacobson 2001; Sinha et al., 2017.

Our detailed answers to Referee #2:

This manuscript describes measurements of aerosol concentrations and optical properties (absorption, scattering, extinction) measured during a month-long period at the Pallas ground site in northern Finland. Filter-based aerosol absorption instruments include two models of aethelometers, a MAAP, a PSAP, and COSMOS. Extinction and scattering coefficients are obtained from CAPS and nephelometer instruments, respectively. Information on black carbon mass and coating thickness is obtained from an SP2, while particle number concentrations are measured by a CPC. In total, this is a comprehensive aerosol measurement suite! The primary focus of the study is to examine the instrument consistency during two time periods – Period 1 is characterized by relatively low aerosol scattering coefficients, while Period 2 sees higher particle scattering coefficients. Aerosol absorption coefficients and number concentrations are similar across both periods, after removing cloud/fog artifacts when ambient visibility was greatly reduced. Backtrajectories are included to provide context for air mass history, which indicate consistently northeasterly winds during Period 2, while the wind directions during Period 1 are much more variable. Despite some degree of variability between the two time periods, the overall aerosol absorption, scattering, and number concentrations are quite low, which would be expected for the Pallas region (far from continental pollution aerosol sources). This challenges many of the instrument comparisons because the measurements are close to the lower limits of detection, and it appears that the data are highly averaged to reduce noise (which is appropriate and fine). Overall, the manuscript is well written, enjoyable to read, and nicely describes the Pallas scientific instruments and the observations obtained during this time period; although, the scientific importance of these observations is not clear from the paper. The depth of analysis is pretty shallow as only a short period of time is being examined and the mean concentrations shown in Table 2 tend to be close to zero (with

large standard deviations relative to the magnitude of the mean), which makes it hard to draw conclusions regarding instrument agreement/disagreement. I'm also unsure if AMT is the appropriate journal for this manuscript. All instruments are commercially available and have previously been described in the literature, and there don't seem to be novel conclusions related to their performance and/or operation that would be informative to the broader scientific community. Since I do not think that the present paper would be acceptable for publication without bringing in additional data and completely rewriting/reframing its scope, I recommend that it be rejected for publication in AMT.

ANSWER: We appreciate your kind words and an excellent summary on our work. We take the message that the manuscript in its previous form is rather confusing and presents no clear value for the community, for which we have completely re-written the results section. Many of your excellent suggestions have been considered in the modified manuscript, and for example the Periods 1 and 2 are no longer separated, and the focus is not on aerosol climatic relevance, but rather on instruments ability to measure in such pristine environments, for which this campaign data serves as a unique data set and can show a broader value. We hope that you find our changes adequate.

Specific Comments: 1) As stated above, the depth of analysis in this paper is low. Simply comparing the data from multiple, filter-based aerosol optical property measurements for a month and reporting summary statistics that largely overlap with zero provides little value regarding the instrument operation or the remote measurement site characteristics. One way to increase the depth of analysis might be to reframe the paper to describe the annual aerosol climatology relevant to the Pallas site. This might include monthly and seasonal data from satellites and/or models that provide some long-term context for the site measurements. It would also be great if more in situ data could be included beyond a single month; although, I recognize that that might be prohibitive. Having more in situ data might allow for more dynamic range in the aerosol abundance and optical properties. Another approach to increase the

depth of analysis might be to examine the Pallas aerosol instrument suite response to laboratory-generated aerosol under very controlled conditions to understand their interconsistency and accuracy. These results could then be compared to the ambient measurements to understand/evaluate the instrument performance. These are just a couple of ideas for the authors' consideration in a potential future manuscript.

ANSWER: Thank you for your excellent suggestion on how to modify the focus of the ms. Finally, we took the decision to present the campaign results in a slightly different context, but with adding no data. We hope that you find this acceptable. One reason is that there are several previous long-term studies made at Pallas and other Arctic sites, but very view studies have focused on the instruments inter-comparisons in pristine field conditions. Secondly, we aim at reporting a laboratory characterization on these very same instruments. However, the results do not fit in this paper and it would change the scope completely, giving very little meaning to present the atmospheric inter-comparison, which is here the main focus. There are previous laboratory studies, too.

2) The sensitivities and lower detection limits given in the abstract (and elsewhere) need to include the averaging interval. I'm assuming that 0.05 Mm-1 as achieved by averaging for > 1 hr. What are the overall instrument accuracies?

ANSWER: Yes, these were added. Defining the accuracy is a question of a reference. Here, we used MAAP as an "absorption reference" and SP2 as a "mass reference", however understanding that these can not provide real references as such. The PSAP provided the best comparison to MAAP.

3) On Line 24, what is meant by the statement, "additional activation of secondary particle formation mechanisms"?

ANSWER: Removed.

4) Strike first sentence on Line 29 as not relevant. The second sentence is referenc-

ing a filter-based measurement method, which I'm assuming is of aerosol absorption yes? Also please correct typos on Line 29 (begun -> began) and Line 31 (graphic ->graphitic).

ANSWER: These were corrected as suggested.

5) On Line 96, it is noted that the Arctic aerosol absorption measurements are "demanding". Is this related to the low concentrations due to a lack of local combustion sources? If so, it might be worthwhile to clarify that, while also noting that such, low-aerosol conditions are also not unique to the Arctic.

ANSWER: This part was re-formulated to refer to pristine environments where techniques suffer from signal-to-noise challenges.

6) Is the mass absorption coefficient (MAC) defined on Line 145 the same as the MAC used on Line 200? On Line 146 it is called a "wavelength dependent specific attenuation".

ANSWER: MAC was removed from line 145, because it is not exactly the same. Thank you for notice.

7) What value(s) of the MAC were applied as mentioned on Line 201?

ANSWER: COSMOS MAC was 8.73 m2 g-1. This was added.

8) On Lines 248 (and elsewhere), I find the extinction-minus-scattering (EMS 1 2) terminology confusing because this is neither an instrument nor a complex method. Rather, it's just a difference between two values. I suggest that it would be worth denoting as σCAPSex$-\sigma$TSI and σCAPSssa$-\sigma$AUR4 to help avoid confusion and to be consistent with the notation in Figure 1.

ANSWER: We are aware that this name has been in use only for a short time, partly because the technique as such is still relatively little applied. However, some previous work (e.g. Modini et al., 2021) use the name "EMS" and we would therefore wish to

keep it as it is, to clarify that the method is the same as in their ms.

9) On Line 257, it is noted that differences in CAPS instrument response may be due in part to "discrepancies in inlet tubing sizes and flow rates", which seems unlikely to me. Can this sentence be clarified to explain how these differences would affect the aerosol measurements?

ANSWER: We agree that the disagreement between the sampling line lengths and tubes was a negligible source of error and is unable to explain much of the difference. As we mention that the nephelometer sampling settings were slightly different, this could have a minor impact. This was re-phased now more clearly in the text.

10) On Lines 274-275, Table 2, and elsewhere, I note that the magnitudes of arithmetic standard deviations often exceed the magnitudes of the arithmetic means, which gives implies non-physical, negative values. Is this because of instrument noise that results in a negative baseline or is this because the observational data are not normally distributed about the mean. At least in the case of the CPC data reported in Table 2, it would seem that the latter explanation is correct, in which case it would be appropriate to report the summary statistics as either a geometric mean */ one geometric standard deviation or as median and percentiles.

ANSWER: You are correct that in some cases data are clearly not normally distributed and the std values lead to unphysical interpretations. We have now preferred the use of median and quartiles values, rather than averages and std in most of the ms tables and figures.

11) Rather than using the qualitative SP2 lag times, can the data be reanalyzed to provide quantitative coating thicknesses? I think that this is important and would help to increase the paper depth of analysis.

ANSWER: In the modified ms version, the aerosol mixing state is no longer discussed since the interpretation of this data (section 3.5) was too uncertain and speculative with

such low concentrations and short campaign time. We hope that you understand this decision.

12) What support is there for the statement made on Lines 310-11 that the more thickly coated aerosols have longer atmospheric ages?

ANSWER: Referring to our previous answer, this section was removed from the modified ms.

13) The final conclusions on Lines 429-434 are not really connected to the data that are presented in this manuscript. What site- and aerosol-specific filter artifacts were presented and overcome? How were good measurement practices and careful data post-processing demonstrated in this study that differ from conventional techniques? To put it bluntly, what is new or novel from the present paper in terms of measurement techniques?

ANSWER: We removed this sentence and aimed at sharpening the conclusions according to the new ms structure, also to answer better “what is new and novel in this paper”.

14) There seem to be too many significant figures presented in Table 3, which imply the absorption coefficient measurements can be made with a precision of 0.001 Mm-1. I realize that the’re a lot of averaging going on here to help tamp down the noise, but I suspect that at least the last digit is probably not significant.

ANSWER: This is probably very much true. We know that these 1h-average values are measured with instruments with a of lower detection limit on the order of $\sim 0.01 - 0.1$ Mm-1. What is the accuracy, is currently unknown since we do not have a reference method for aerosol absorption.

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